Large-scale equatorward transport of ozone in the subtropical lower stratosphere

Erik C. Richard,[1] Kenneth C. Aikin,[1] and Eric A. Ray[1]
Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado, USA

Karen H. Rosenlof and Thomas L. Thompson
Aeronomy Laboratory, National Oceanic and Atmospheric Administration, Boulder, Colorado, USA

Andrew Weinheimer, Denise Montzka, David Knapp, Brian Ridley, and Andrew Gettelman
National Center for Atmospheric Research, Boulder, Colorado, USA

Received 18 June 2003; revised 19 August 2003; accepted 27 August 2003; published 2 December 2003.

[1] Anomalous vertical profiles of ozone were observed in the subtropical lower stratosphere (near south Florida) in July 2002 during the NASA-sponsored Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus Experiment measurement campaign. It is shown that there is an enhancement of ozone (initially >150%) above the tropopause extending up to ~410 K potential temperature. This ozone increase is the result of recent transport of middle-and high-latitude lower stratospheric air into the subtropics. This meridional transport was a consequence of a geostrophic flow pattern established by a quasi-stationary anticyclone centered over the south central United States that persisted for much of July 2002. We show the spatial and temporal extent of meridional isentropic transport into the subtropics by examining the ozone vertical profiles in combination with the NO_y:O_3 correlations as well as isentropic back trajectory calculations. The anomalous ozone profiles are also reproduced in a global chemical transport model. *INDEX TERMS:* 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); *KEYWORDS:* ozone, lower stratosphere, large-scale transport

Citation: Richard, E. C., K. C. Aikin, E. A. Ray, K. H. Rosenlof, T. L. Thompson, A. Weinheimer, D. Montzka, D. Knapp, B. Ridley, and A. Gettelman, Large-scale equatorward transport of ozone in the subtropical lower stratosphere, *J. Geophys. Res.*, *108*(D23), 4714, doi:10.1029/2003JD003884, 2003.

1. Introduction

[2] The exchange of air between the midlatitudes and the tropics in the upper troposphere (UT) and lower stratosphere (LS) can alter the chemical composition near the tropopause and impact the chemical and radiative balance of the atmosphere [*Houghton et al.*, 2001]. This is particularly true for the transport of Northern Hemisphere (NH) midlatitude air into the tropical ascent region. The potential for polluted NH air to reach the tropical upwelling region is of concern because of serious impacts on the chemistry of the stratosphere and the balance of ozone [*Stolarski et al.*, 1995; *World Meteorological Organization*, 2003]. In order to assess the long-term impacts of both anthropogenic and natural emissions on the chemical balance of ozone, reliable estimates of the magnitude of meridional exchange between the midlatitudes and the tropics are necessary.

[3] In the UT region in particular, NH subtropical high-pressure systems (anticyclones) have been shown to affect the circulation patterns in the LS and thus influence the long-range transport of chemical constituents. *Dunkerton* [1995] investigated the climatology of large-scale circulations adjacent to monsoon regions in the NH summer. The results of this study showed that the horizontal circulations from quasi-stationary anticyclones over North America can penetrate into the LS up to ~25 km and are effective in long-range meridional transport of chemical constituents. Observational evidence for this was seen in the poleward transport of Mt. Pinatubo aerosol. Using SAGE-II aerosol extinction data, *Trepte et al.* [1993] showed that significant LS aerosol transport into the northern mid and high latitudes was directly associated with UT quasi-stationary anticyclonic circulation systems during the NH summer.

[4] Long-range transport associated with synoptic-scale disturbances can also directly affect the distribution and variability of ozone abundance in the UT/LS. In the LS, below 25 km, the photochemical lifetime of ozone is several months to a year [*Solomon et al.*, 1985; *Avallone and Prather*, 1996]. Since this is generally much longer than

[1]Also at Aeronomy Laboratory, National Oceanic and Atmospheric Administration, Boulder, Colorado, USA.

0148-0227/03/2003JD003884$09.00

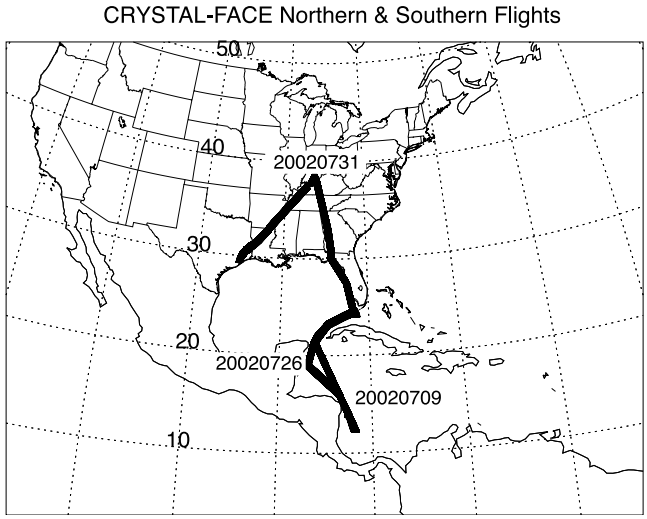

Figure 1. WB-57F flight tracks for northern survey transit flight (20020731) and two southern survey flights (20020709 and 20020726). The total latitude coverage here spans from 39°N down to 12°N.

transport timescales, transport processes primarily determine the ozone distribution and variability in the tropical to midlatitude lower stratosphere (excluding the high latitudes in late winter/early spring). Consequently, ozone distributions are dependent on both latitude and season and have been used extensively to infer transport in the lower stratosphere [*Reid and Vaughan*, 1991; *Vaughan and Timmis*, 1998; *Rood et al.*, 2000; *Proffitt et al.*, 2003].

[5] Evidence suggests that changes in ozone abundance in the lower stratosphere, particularly near the tropopause, can have potential climate impacts. For example, several modeling studies have examined the effect that changes in the vertical distribution of ozone have on radiative climate forcing [*Ramaswamy et al.*, 1992; *Lacis et al.*, 1990; *Forster and Shine*, 1997]. Radiative-convective models show that, as a function of height, ozone changes near the tropopause have the largest influence on surface temperature. Therefore, depending on the spatial extent, changes in ozone abundance near the tropopause can cause significant perturbations in the direct radiative forcing and thus influence climate in the troposphere.

[6] The ozone vertical distribution in the lower stratosphere near the tropopause, particularly in the tropics and subtropics, is difficult to characterize accurately from satellite based instruments. Typically, these instruments rely on an atmospheric backscattering technique and are most sensitive to ozone concentrations above 25 km. Furthermore, long-term ozone monitoring and ozonesonde measurement stations are located predominantly in northern midlatitudes with few located throughout the southern midlatitudes and even fewer at tropical latitudes [*Logan*, 1994, 1999].

[7] In this paper we report on extensive in situ measurements made in the summer subtropical UT/LS, primarily between 340 and 440 K potential temperatures, during the NASA Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus Experiment (CRYSTAL-FACE) campaign in July 2002. During this time period, a persistent quasi-stationary anticyclone was centered over the south central United States. Fast response in situ sampling of this region provided a unique opportunity to accurately measure the ozone distribution in the UT/LS region adjacent to and downstream of a large anticyclone. Furthermore, the simultaneous measurements of trace and reactive gases as well as aerosols and meteorological data add a powerful complement to allow independent quantification of chemical and dynamical processes. Therefore the measurements taken during the CRYSTAL-FACE campaign add significantly to the body of ozone data in the tropics and subtropics.

2. Measurements

[8] The in situ measurements reported here were acquired from fast response instruments on board the NASA WB-57F high-altitude aircraft. These measurements included O_3 [*Proffitt and McLaughlin*, 1983], CH_4 [*Richard et al.*, 2002], NO_y [*Ridley et al.*, 1994], and pressure and temperature [*Thompson and Rosenlof*, 2003]. During the CRYSTAL-FACE mission, a total of 14 science flights were completed with the NASA WB-57F out of Boca Chica NAS located near Key West, Florida (24.6°N, 81.7°W). The flights spanned the entire month of July starting on June 29th (transit) and ending on the 31 July 2002 (return transit). The latitude coverage for these flights ranged from 39°N down to 12°N. Figure 1 shows the latitude coverage for the northernmost (20020731 return transit) and the two southernmost (20020709 and 20020726) flights (following format convention, the flight date is written as yyyymmdd). The two southern survey flights followed similar flight profiles, occurred approximately 3 weeks apart and covered ~12° of latitude. The altitudes covered the region from the UT to the LS up to 450 K (70 hPa).

[9] Additionally, during the CRYSTAL-FACE campaign there were many southern Florida flight profiles designed to sample regions around convection. All of the flights involved numerous stepped profiles between the UT and LS up to ~19 km altitude (460 K). Overall, the CRYSTAL-FACE WB-57F flights in the UT/LS allowed for sufficient characterization of air masses over extended times and latitudes.

3. Results and Discussion

3.1. Data Overview

[10] An example of the UT/LS in situ flight data is shown in Figure 2. Plotted here are a time series of in situ O_3, CH_4 and NO_y (the sum of total reactive nitrogen species; NO_y = $NO + NO_2 + NO_3 + 2N_2O_5 + HNO_3 + ClONO_2 + HO_2NO_2$ + peroxyacetal nitrate (PAN)+...) measurements recorded simultaneously on 20020723. The altitude profile for this 1 hour and 40 min section of the flight includes several stepped legs going from ~200 hPa (~345 K) to 110 hPa (385 K).

[11] This section of the flight highlights several interesting features. The chemical constituent data in this flight section reveal a very sharp transition layer seen clearly at ~120 hPa (377 K) located in the flight track between 80000 and 81000 UT seconds. This is evident in the O_3 and tracer mixing ratios as a function of pressure. In this region, as the WB-57F climbed from 150 hPa (354 K), just above the

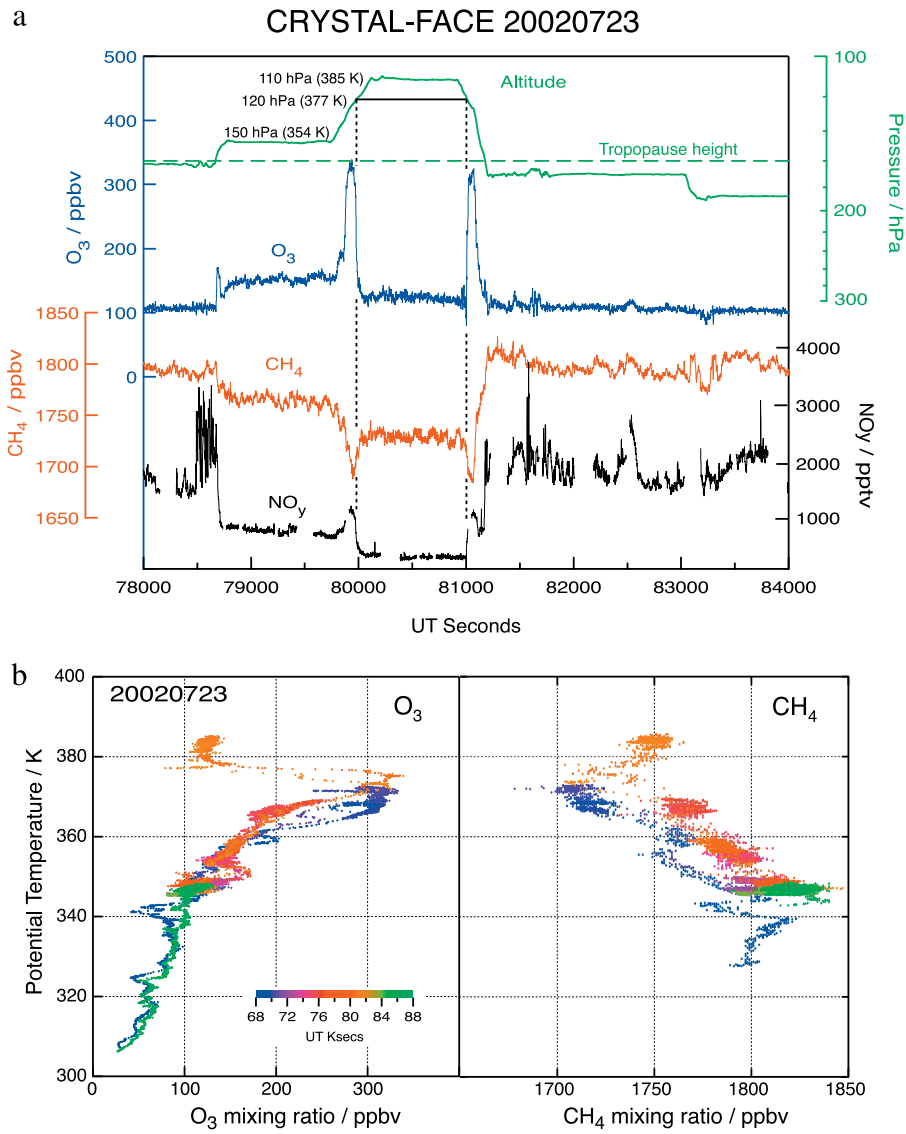

Figure 2. (a) Flight track traces for pressure, O_3, CH_4, and NO_y for a 100 min section during the flight of 20020723. The green dashed line represents the Key West thermal tropopause as measured by sonde. (b) Vertical mixing ratio profiles for O_3 and CH_4 as a function of potential temperature for 20020723.

local tropopause, to 110 hPa (385 K), the ozone mixing ratio increases rapidly as expected; ozone is generally a proxy for altitude in the lower stratosphere. However, at 120 hPa, the ozone mixing ratio decreases sharply from 350 ppbv to ~100 ppbv. Above 120 hPa the ozone remains relatively constant at ~100 ppbv. Upon descent at ~81000 UT seconds, the layer is again encountered at 120 hPa showing the rapid rise to >300 ppbv ozone mixing ratio and then decreases as altitude decreases.

[12] Close examination of other tracer data also shows this interesting layer. Coincident with the ozone gradients, two sharp, anticorrelated transitions occur in both the CH_4 and NO_y mixing ratios suggesting air of different origins. Together with ozone, NO_y and CH_4 help determine an air parcel's history. In the lower stratosphere, both NO_y and O_3 have positive vertical gradients in mixing ratio because of similar source/sink relationships. In contrast, CH_4 is a tropospheric source gas with a negative vertical gradient in the stratosphere. Therefore, for stratospheric air, O_3 mixing ratios will be high in conjunction with higher NO_y and lower CH_4 mixing ratios; the reverse is true for tropospheric air.

[13] Figure 2b shows the vertical distribution of the in situ O_3 and CH_4 data for the flight of 20020723 plotted as a function of potential temperature calculated from the in situ pressure and temperature measurements. The sharp transition gradient can be seen clearly between 375 and 380 K potential temperature (at 120 hPa in Figure 2a). Within this narrow potential temperature region the ozone changes by 200 ppbv (>150%). Examination of the CH_4 data in this same region shows a noticeable decrease in mixing ratio (~50 ppbv decrease) coincident with the O_3 increase and is consistent with a greater percentage of photochemically aged stratospheric air.

[14] The in situ LS data shown in Figure 2 is indicative of a layered structure of "younger" air above 120 hPa (377K),

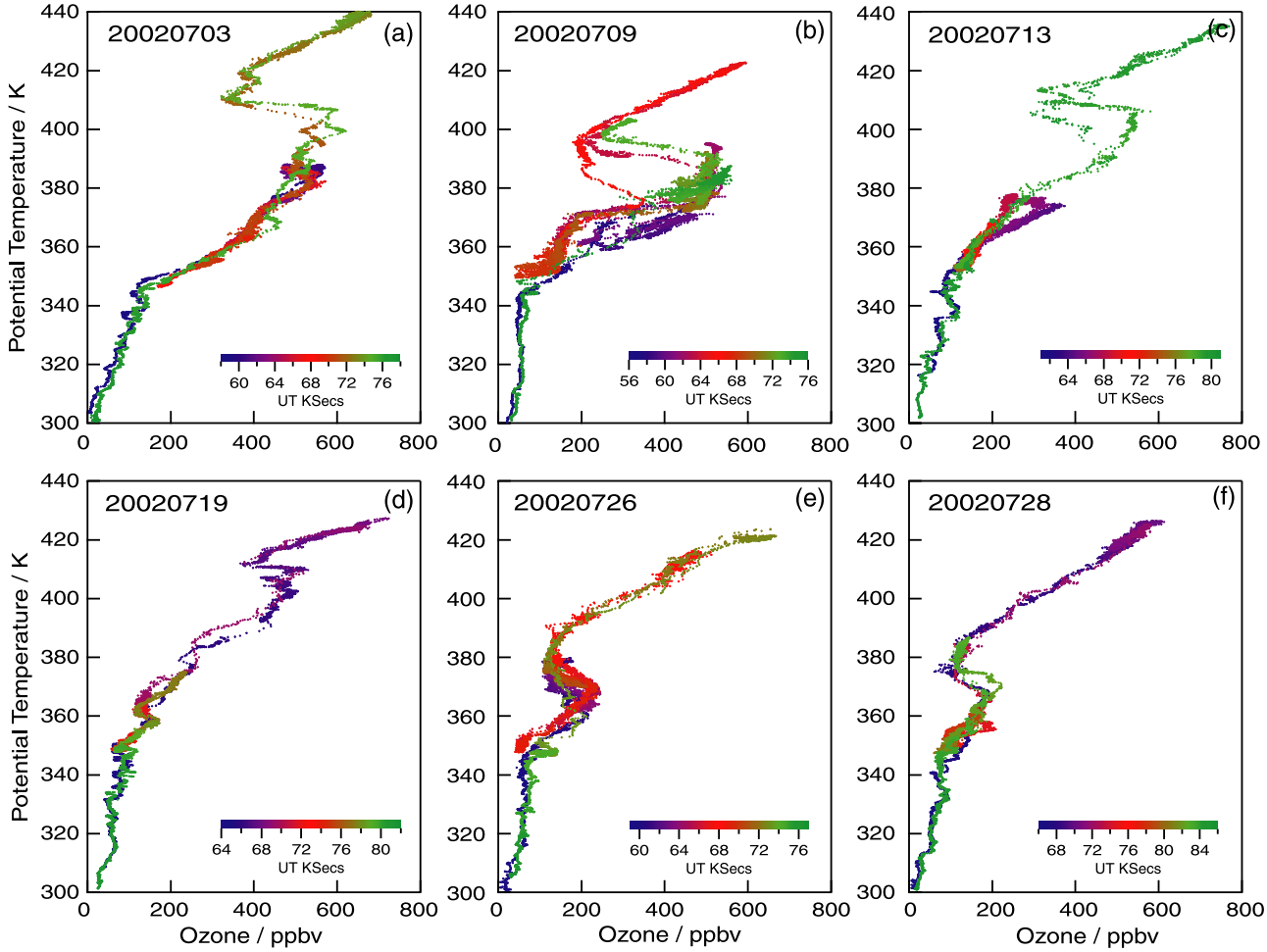

Figure 3. Vertical ozone mixing ratio profiles as a function of potential temperature for several CRYSTAL-FACE flights during July 2002.

showing lower O_3 and NO_y and higher CH_4 mixing ratios, and "aged" stratospheric air below corresponding to higher O_3 and NO_y and lower CH_4 mixing ratios. Additionally, the in situ water vapor data above 120 hPa (not shown) remains low at ~5 ppmv, consistent with dehydration of the younger tropical air as a result of recent upward transport through the cold tropical tropopause [*Kley et al.*, 1982]. In the lower stratosphere, this type of layered structure is observed throughout all the flights of the CRYSTAL-FACE mission.

3.2. Ozone Vertical Profiles

[15] Figure 3 shows the vertical profiles of ozone as a function of potential temperature for several of the science flights spanning the July 2002 duration of the CRYSTAL-FACE campaign. Generally, there is sufficient altitude profiling to cover the UT/LS region with most flights covering a potential temperature range from 300 to 440 K.

[16] As shown in the previous section, a characteristic feature clearly evident in all of the profiles is a localized region of large vertical gradients in ozone mixing ratio just above the local tropopause occurring between approximately 350 and 410 K potential temperature. Examination of the vertical profiles shows that the gradient regions decrease over time both in magnitude and altitude. Initially, the ozone differences are ~300 ppbv localized near 400 K. By the end of the mission, the region only spans from approximately 345 to 370 K with ozone differences of ~100 ppbv. The profiles show that over time the ozone mixing ratio changes occur predominantly in the 340–400 K region. Note that above ~400 K in all of the profiles the vertical gradient in O_3 mixing ratio does not vary considerably over the extent of the measurements throughout July. The data suggest an ozone enhancement between the local tropopause and ~400 K initially on 20020703, decreasing to ~370 K by 20020728. This structure is indicative of a persistent, large-scale intrusion of elevated O_3 into the subtropical LS.

[17] To further confirm that this is a positive ozone anomaly we can compare the profiles with a "climatology" for the time and region. In Figure 4 we compare a summer 2002 vertical O_3 profile with that of a previous summer profile of similar location. Shown here is the ozone vertical profile as a function of potential temperature between 300 and 460 K for the flight of 20020703. Also shown on Figure 4 is the August average ozone data recorded by the NOAA in situ ozone instrument on board the NASA ER-2 during ascents and descents at Jacksonville, Florida (30.5°N, 81.7°W) during the CAMEX-IV mission in late summer 2001 (approximately 1 year earlier). Between these two summers, the data show that above 410 K there is fairly good agreement with both the absolute value

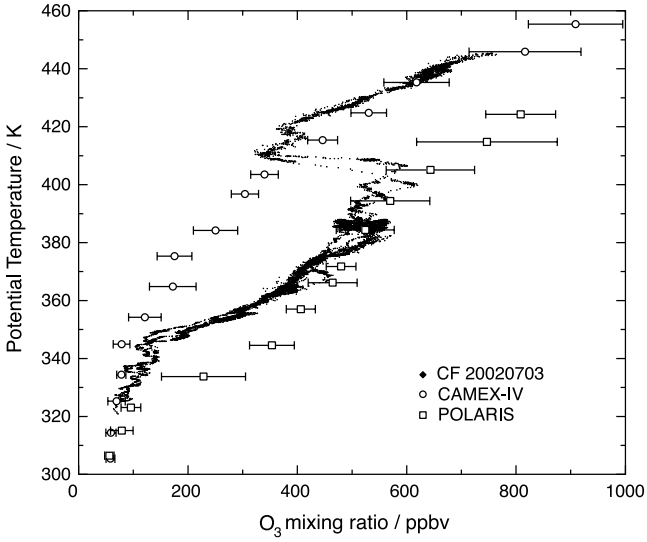

Figure 4. Comparison of ozone mixing ratio vertical profile as a function of potential temperature between CRYSTAL-FACE 20020703 (dots), CAMEX-IV August 2001 average (circles) and high-latitude POLARIS July 1997 average (squares). The error bars correspond to the 1-σ standard deviation of the averaged ozone mixing ratio.

and the vertical gradient in ozone mixing ratio, suggesting this region during July 2002 to be more typical of the LS in this midlatitude/subtropical region. Therefore we suggest the observed ozone gradients are the result of a large enhancement of ozone mixing ratio between ~350 K (just above the local tropopause) and 410 K potential temperature.

[18] Interestingly, as an additional comparison we also show in Figure 4 the July high-latitude average for in situ ozone data from the summer 1997 POLARIS mission for profiles from Fairbanks, Alaska (64.8°N, 147.6°W) [*Newman et al.*, 1999]. Here the POLARIS high-latitude ozone data agree well with the ozone profile from CRYSTAL-FACE between 360 K and 400 K. This seems to suggest that the enhancement of ozone between the 350 K and 410 K levels is due to the meridional transport of higher latitude air into the subtropical LS. This will be investigated further in the following sections.

3.3. Ozone Spatial and Temporal Variability

[19] There were two flights during the mission that allowed for a direct study of the ozone spatial and temporal variability in the UT/LS. The flights of 20020709 and 20020726 were southern survey flights covering a latitude range from Key West (24.6°N) to 12°N and 14°N, respectively. These flights followed nearly identical flight tracks and provided a unique opportunity to characterize the time evolution of the O_3 meridional distribution over 12° of latitude in the subtropical UT/LS.

[20] Figures 5a–5d show the latitudinal dependence of the ozone vertical profiles in the UT/LS for 20020709 and 20020726 between 25°N and 12°N. These profiles are grouped into four latitude regions each spanning ~3° of latitude. A comparison of the early and late O_3 vertical profiles within each latitude region shows clearly the spatial and temporal variation of the ozone mixing ratio between 360 and 400 K. Comparison of the 20020709 profiles between the different regions shows that the intrusion weakens with decreasing latitude. This weakening occurs predominantly from the higher layers. By the flight of 20020726, there are just small remnants of the intrusion, located in the small altitude region between the tropopause and ~370 K. Note, however, that there is much less latitudinal dependence in the profiles on the 26th as compared to the 9th. Because of these differences, this study will focus on data from these two flights. Both back trajectory calculations and chemical tracer correlations will be used to try to understand the air mass differences causing this behavior.

3.4. Trajectory Analysis

[21] Figures 6a and 6b show the meteorological situation for the first and last weeks, respectively, in July over most of the North American continent. Plotted is geopotential height on the 100-hPa pressure surface. This height is at 380–400 K potential temperature in the subtropics and represents the situation in the LS. Figure 6a shows the weekly average for the time period of 20020701–20020708. Here anticyclonic flow around a blocking high located over the central US produced equatorward flow over the eastern United States. The weekly average in Figure 6b shows that by the last week in July the high has weakened, with more zonal flow apparent in the LS.

[22] A series of 5-day isentropic back trajectories was run in an effort to try to understand the air mass differences, both vertically and temporally, between 20020709 and 20020726. The meteorological fields used for the back trajectory runs are from the National Centers for Environmental Prediction (NCEP) aviation assimilated model on 1° × 1° horizontal resolution. Figures 7a–7d show the isentropic trajectories initialized on the 420 K and 380 K surfaces at 23°–27°N for 20020709 and 20020726. The 420 K and 380 K potential temperatures were chosen to highlight the regions showing minimum and maximum differences in the observed ozone mixing ratios, respectively, between the two southern survey flights.

[23] It can be seen in Figures 7a and 7c that at 420 K both the 9th and the 26th show an easterly flow of LS midlatitude and subtropical Atlantic air with very little meridional transport. However, comparison of Figures 7b and 7d shows that at 380 K there is a dramatic difference in air mass history between the 9th and 26th. On the 9th, the flow pattern at 380 K is influenced strongly by the quasi-stationary anticyclone producing large equatorward flow from higher latitudes. In contrast, by the 26th the flow at 380 K is similar to that observed at the higher potential temperatures showing predominantly easterly and southeasterly flow of subtropical air. It is also interesting to note the similarity between the 380 K flow pattern on the 26th (Figure 7d) with that of the 420 K flow pattern on the 9th (Figure 7a). This corresponds to a shift to a more tropical summertime regime over south Florida. There is also an increase in tropopause altitude over the month of July, which would also indicate a shift to a more tropical regime.

[24] On the flight of 20020709, the strength of the anticyclone and its influence on the meridional transport at 380 K is observed even at the southernmost latitudes.

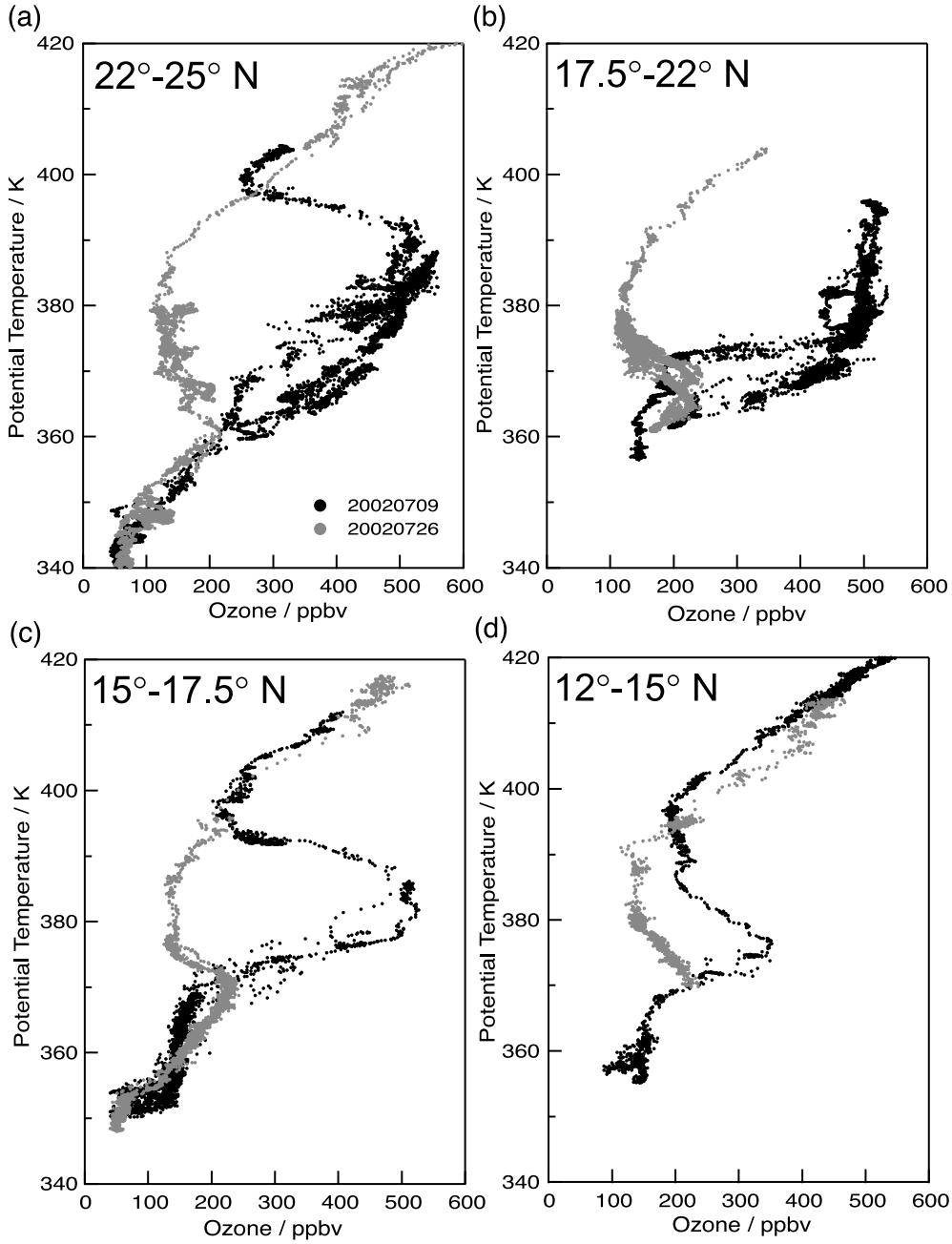

Figure 5. Comparison of the vertical profiles of ozone mixing ratio as a function of potential temperature between the southern survey flights of 20020709 and 20020726 for (a) 22°–25°N, (b) 17.5°–22°N, (c) 15°–17.5°N, and (d) 12°–15°N.

Figure 8 shows the 5-day back trajectory on the 380 K surface for 20020709 initialized in a latitude grid of 12°–16°N (southernmost part of flight track). As can be seen, although somewhat weaker than observed at higher latitudes (compare Figure 7b), the 12°–16°N region is still influenced by meridional flow from the midlatitudes. Again, this is consistent with the observations showing smaller ozone mixing ratio enhancements at 380 K in the 12°–15°N region (compare Figure 5d).

3.5. NO_y:O_3 Correlation

[25] As a further test of the extent of the high to midlatitude advection into the subtropics, we can use ozone tracer correlations that show distinct relationships as a function of latitude. It has been well established that for lower stratospheric long-lived trace gases both spatial and temporal variations in mixing ratios are well correlated [*Kelly et al.*, 1989; *Strahan et al.*, 1989; *Proffitt et al.*, 1990, *Plumb and Ko*, 1992; *Proffitt et al.*, 2003]. Therefore examination of tracer-tracer relationships in addition to the individual profiles provides a more rigorous test of meridional transport [*Murphy et al.*, 1993; *Volk et al.*, 1996; *Tuck et al.*, 1997; *Herman et al.*, 1998; *Flocke et al.*, 1999].

[26] A powerful diagnostic for transport processes in the lower stratosphere is the correlation of ozone with NO_y. Previous extensive in situ aircraft measurements have

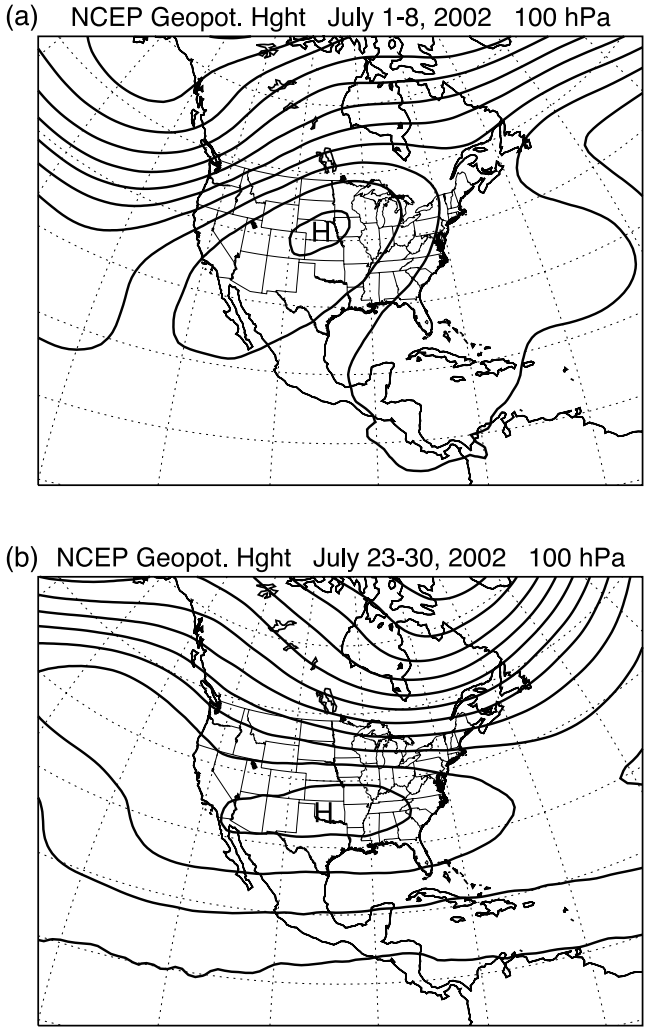

Figure 6. Plot of 8-day average geopotential height on the 100 hPa surface for (a) 1–8 July 2002 and (b) 23–30 July 2002.

shown a strong positive correlation between NO_y and O_3 abundance between the tropopause and 21 km [*Murphy et al.*, 1993; *Fahey et al.*, 1996]. In this region, the correlation is sensitive to transport and mixing processes since both species act largely as long-lived tracers separated from source regions at higher altitudes and sink regions at the tropopause and below. In particular, the slope of the NO_y to O_3 correlation (or alternatively, the NO_y/O_3 ratio) is found to be nearly independent of altitude but significantly dependent on latitude. Therefore the slope of the correlation emphasizes horizontal chemical gradients in the lower stratosphere.

[27] Throughout the midlatitudes, the NO_y/O_3 ratio in the lower stratosphere is found to be $\sim$0.003 [*Murphy et al.*, 1993]. The reason for this can be attributed to fast quasi-horizontal mixing occurring in regions absent of significant sources and sinks for either NO_y or O_3. In the inner tropics, however, the ratio decreases to a value of less than half the midlatitude value due to O_3 production in the lower stratosphere, whereas at higher latitudes ($\geq$50°N) the observed NO_y/O_3 ratio values in the lower stratosphere increase substantially. The increase of NO_y/O_3 in the high-latitude lower stratosphere is indicative of air that is photochemically aged where NO_y production has occurred via N_2O photolysis.

[28] Figure 9 shows the NO_y:O_3 correlations for both 20020709 and 20020726 as a function of potential temperature in the altitude region above (Figure 9a) and within (Figure 9b) the ozone enhancement. It should be noted here that the NO_y data have been filtered to include only gas phase NO_y. This is mainly a concern for data near the tropopause, where NO_y can be condensed on cirrus, and should not affect the data for the LS region. To filter the data, we use the total surface area volume data from the CAS particle instrument. As a conservative estimate we use only NO_y data when total particle surface area volume is less than 50 μm^2 cm^{-3}. (D. G. Baumgardner, personal communication, 2002).

[29] As shown previously, the region above 400 K represents air above the strong meridional transport region. Figure 9a shows all of the NO_y:O_3 data for both flights above 400 K agree very well with the 3:1000 midlatitude line. Careful examination shows that most of the data lie just slightly below the line, particularly for 20020709. Previous in situ measurements in the LS have shown that in the tropics the NO_y/O_3 ratio decreases sharply to $\sim$0.001 with the transition region occurring in just a few degrees of latitude [*Fahey et al.*, 1996]. Therefore this slightly lower ratio above 400 K may be suggestive of a small tropical influence. This can be seen in the trajectory result for this region showing a small southeasterly contribution in contrast to the strong easterly flow for the 26th. Interestingly, the overall agreement with the midlatitude NO_y/O_3 ratio of 0.003 holds even at the southernmost latitudes (12°N) indicating that this tropical transition region in this location is south of 12°N latitude during this time period.

[30] The NO_y:O_3 correlation for the region encompassing 360–400 K is shown in Figure 9b. This corresponds to the potential temperature region of the ozone enhancement. Here all data from the 9th and the majority of data from the 26th have NO_y/O_3 ratio values substantially greater than 0.003 (i.e., lie well above the 3:1000 midlatitude line). Note, however, that although it is more scattered there is still an overall positive correlation between NO_y and O_3.

[31] What stands out clearly in this potential temperature region for 20020709 is the significantly elevated NO_y between 400 and 550 ppbv ozone. Here, however, the elevated NO_y is anticorrelated with O_3. Although we will not focus on this here, we note that H.-J. Jost et al. (Observations of mid-latitude forest fire plumes deep in the stratosphere, submitted to *Science*, 2003, hereinafter referred to as Jost et al., submitted manuscript, 2003) and *Hudson et al.* [2003] have examined this elevated NO_y region with respect to CO and particle composition, respectively. The results of their studies suggest that there is a significant midlatitude biomass burning influence in the subtropical LS. Evidence for these biomass burning signatures are seen in isolated regions throughout the subtropical flights and occur predominantly in the 340 to 380 K region with very little influence above 400 K.

[32] The general features of the NO_y:O_3 correlation in Figure 9b are in agreement with the previous conclusions,

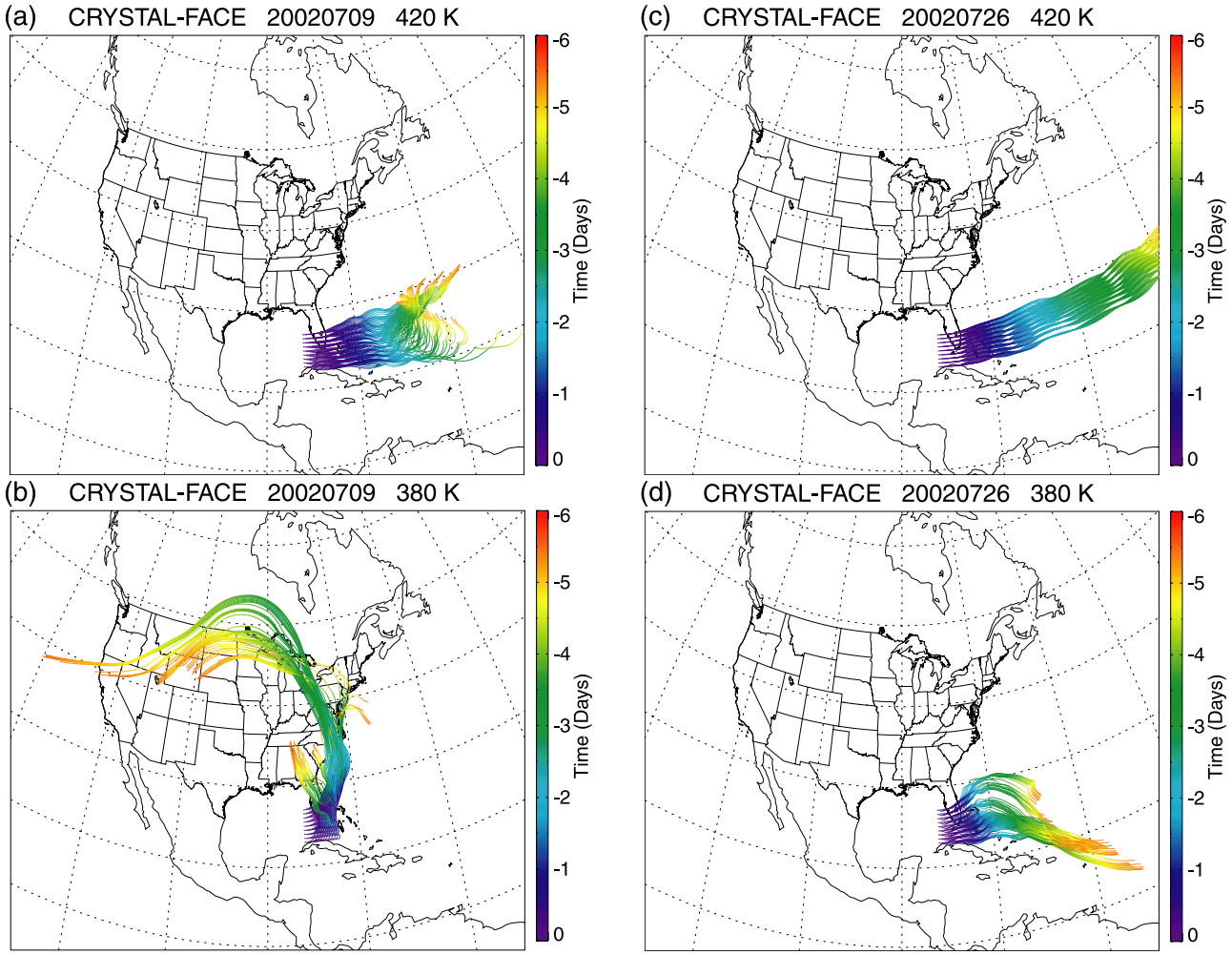

Figure 7. The 5-day isentropic back trajectories for 20020709 at (a) 420 K and (b) 380 K and for 20020726 at (c) 420 K and (d) 380 K.

namely, that relatively high-latitude LS air has been transported equatorward over an extended period of time. The presence of larger NO_y/O_3 ratios in the lower stratosphere at high latitudes is indicative of air parcels that have remained longer or traveled higher in the stratosphere (i.e., photochemically aged air). Previous measurements in the UT/LS at high latitudes indicate higher ratios, typically found to be 0.004–0.006 [*Murphy et al.*, 1993; *Weinheimer et al.*, 1993, 1994]. For example, this larger NO_y/O_3 ratio can be seen in the summer northern latitude LS data from the POLARIS mission based out of Fairbanks, Alaska, in 1997.

[33] Comparing the data between the 9th and the 26th for this lower altitude region, we see that over time the air is beginning to approach a more typical midlatitude condition (i.e., NO_y/O_3 = 0.003), consistent with the vertical profiles and trajectory results. By the flight of 20020726, all of the NO_y in the 360–400 K region corresponds to O_3 mixing ratios less than 250 ppbv. This suggests that over the time period of the measurements, the anticyclonic circulation weakened and allowed the reestablishment of zonal flow in this region causing an increase in the fraction of midlatitude air near southern Florida.

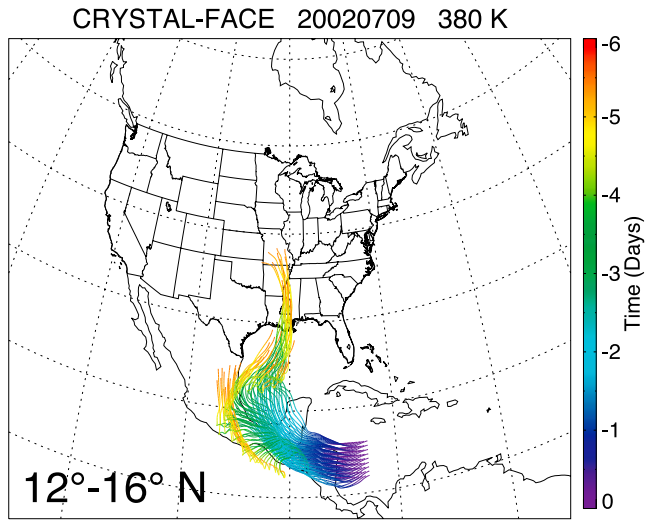

Figure 8. The 5-day isentropic back trajectory at 380 K for 20020709 initialized at 12°–16°N.

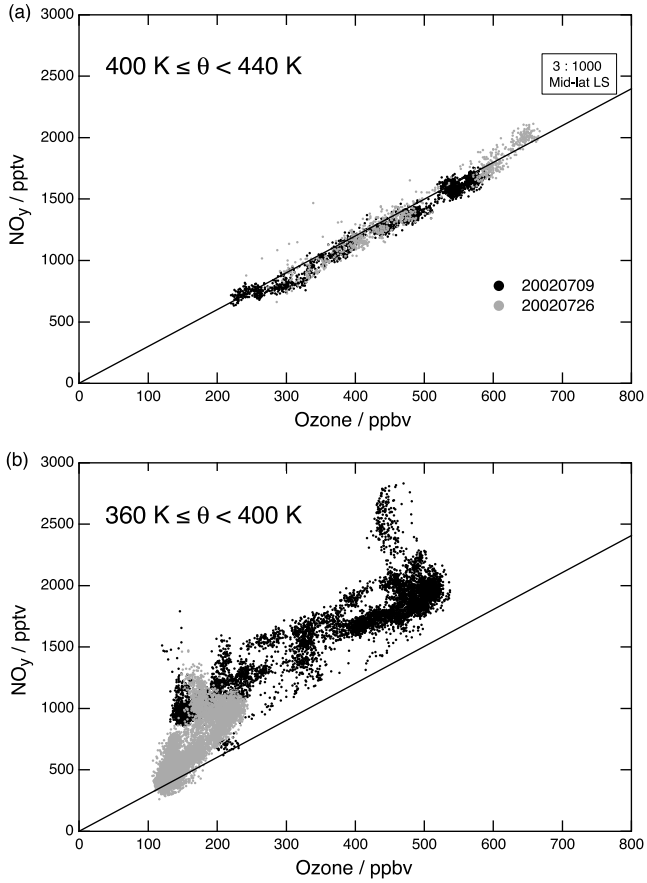

Figure 9. Correlation scatterplot for NO_y versus O_3 for southern survey flights of 20020709 and 20020726 for the potential temperature intervals of (a) 400–440 K and (b) 360–400 K.

3.6. Origin of the Ozone Anomaly

[34] Of interest is from how far north the ozone anomaly between 350 and 400 K originated. Indications from both the O_3 vertical profiles and the NO_y:O_3 correlations are that this air is from latitudes comparable to that sampled during the POLARIS experiment based in Fairbanks, Alaska. Examination of the profile as a function of potential temperature (Figure 4) shows that ozone above 420 K is in good agreement with that sampled during the CAMEX-IV experiment in Florida the year before, while that below 360 K is a mixture of high-latitude and midlatitude ozone values.

[35] To see how the anomalous CRYSTAL-FACE profiles compare with a more complete climatology, we have constructed average ozone on a potential temperature grid using data from the Halogen Occulation Experiment (HALOE) flying on the Upper Atmosphere Research Satellite (UARS) [*Russell et al.*, 1993]. This climatology was done for June, July, and August for the years 1993–2002. A comparison of the climatological profiles with the flight of 9 July 2002 is shown in Figure 10. This comparison also indicates that the bulk of the air at the peak of the anomalous ozone must have originally resided at polar latitudes, likely above 60°N. The transport from the north had to be relatively rapid, as there is no evidence in the profile taken near Key West of dilution from mixing with surrounding midlatitude air between 380 and 390 K. Back trajectories indeed show transport from 50°N within 5 days (See Figure 7b). Even as far equatorward as 13°N, the air below 380 K has high-latitude characteristics.

[36] This type of equatorward transport has the potential to bring air with middle-and high-latitude characteristics into the main uplift region that serves as the source of overworld air. In this particular case, there were also traces of tropospheric pollutants, as indicated by the presence of aerosols and high CO with biomass burning signatures (Jost et al., submitted manuscript, 2003). It also has the potential to bring relatively high water vapor air into the low-latitude rising region, thereby bypassing the tropical cold trap. Long-term changes in the flow that affect the frequency of these sorts of events could be another possibility for the observed stratospheric water vapor changes [*Oltmans et al.*, 2000; *Rosenlof et al.*, 2001].

[37] The limited data from the aircraft do not allow a determination of whether the perturbed ozone air mass mixes irreversibly into the tropics. However, assimilated data trajectory calculations [*Tuck et al.*, 1997] do show that over 40% of the air in the tropical band from 10°S to 10°N below 400 K in the fall season originated 2 months earlier from poleward of 20°N. There is clearly the potential for these sorts of transport events to impact the composition of the inner tropical lower stratosphere.

[38] Transport across the subtropical jet near the tropopause associated with flow around anticyclones has been noted previously by *Postel and Hitchman* [1999]. In their study, they examine the statistics of Rossby wave breaking on the 350 K potential temperature surface near the subtropical jet. They note the highest frequency of these wave breaking events (and induced irreversible transport) occurs immediately downwind of summertime anticyclones. This appears to be the mechanism acting in this particular situation. Although the localized aircraft measurements do not give a good idea of the longitudinal scales involved,

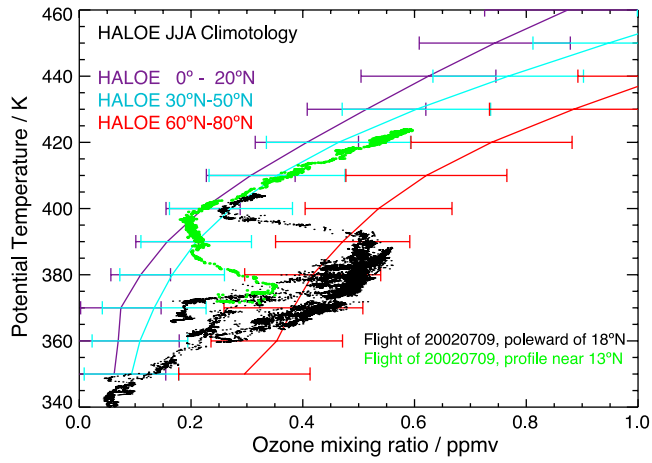

Figure 10. Summer (JJA) ozone climatology for the years 1993–2002 using data from HALOE. These are shown for the tropics/subtropics (purple curve), the midlatitudes (blue curve), and high latitudes (red curve). Also shown are the ozone distributions for 20020709 poleward of 18°N (black dots) and for 20020709 near 13°N (green dots).

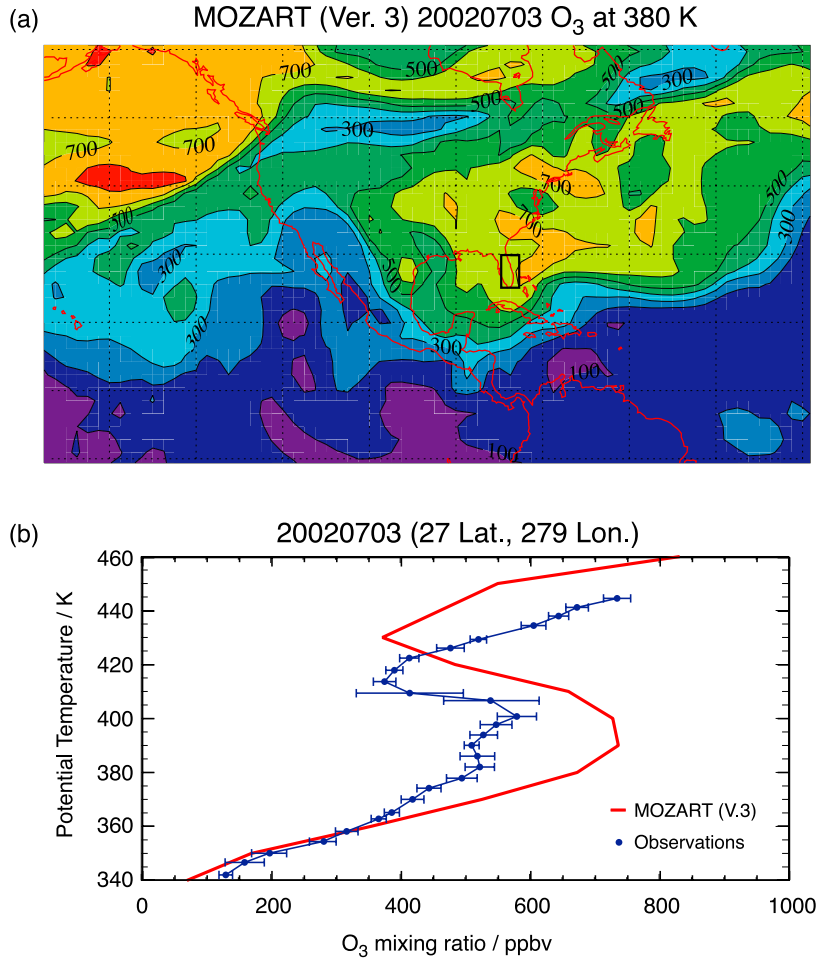

Figure 11. (a) MOZART 3 ozone field averaged for 3 July 2002 interpolated to the 380 K potential temperature surface. The plot is from 0° to 60°N latitude and from 180°W to 30°W longitude. Contour spacing is 100 ppbv. The rectangle indicates the location of the profile in the bottom plot. (b) Profile of ozone (red curve) from MOZART 3 on 3 July 2002 at 27°N, 81°W (indicated by the rectangle in Figure 11a). Model has been interpolated to potential temperature as a vertical coordinate. Also shown is the observed ozone profile for 3 July 2002 (solid blue circles). Each circle represents the 4 K average of the ozone data, and the error bars correspond to the 1-σ standard deviation of the ozone mixing ratio within that 4 K interval.

examination of the NCEP reanalysis for the month of July (not shown) indicates that flow early in the month went around the anticyclone, then entered a breaking wave induced mixing region extending from Florida out into the Atlantic. The anticyclonic flow and associated downwind mixing are largely gone by mid-month, but some signature in the ozone remains even on the flight of the 28th (see Figure 3f), indicating that some midlatitude ozone has been irreversibly transported into the tropics.

[39] The limited data available from the aircraft experiments does not allow a quantification of the mass or ozone flux due to this event. Three-dimensional model analysis will be needed, but first we need to demonstrate that such a model reproduces this type of event.

4. Three-Dimensional Chemical-Transport Model Comparison

[40] In an effort to elucidate the larger-scale features of the meridional transport observed here, we compare the observations with the results of a high-resolution 3-D chemical transport model: Model for Ozone and Related Chemical Tracers (MOZART), version 3 [*Brasseur et al.*, 1998]. The model is driven by European Centre for Medium-Range Weather Forecasts analyzed meteorological fields and is run at T63 resolution (1.9° × 1.9°) in the horizontal with 60 levels in the vertical from the surface to 65 km. This corresponds to 1.2 km vertical resolution in the UT/LS. The model was initialized on 1 June 2002 from a model climatology derived using GCM winds. The model includes 50 chemical species, with approximately 150 photochemical and chemical reactions. For comparison, the model output has been interpolated to isentropic surfaces.

[41] Figure 11a shows the results of the MOZART (version 3) ozone field averaged for 3 July 2002 at 380 K. A large region of enhanced ozone is observed extending from the Midwest of the United States to the western Atlantic with the highest mixing ratios found along the east coast line and east of Florida. Figure 11b shows a compar-

ison between the modeled and observed vertical mixing ratio of ozone between 340 and 460 K potential temperature for 3 July 2002. The modeled profile represents the vertical distribution at 27°N and 81°W (the region shown by the rectangle in Figure 11a) and encompasses the region sampled by the WB-57F on the same day. As can be seen here, the in situ ozone data is well represented by the MOZART model run. The location of the maximum ozone enhancement between 380 and 400 K is reproduced; however, the magnitude of this enhancement is overestimated by the model. Above 420 K the model underestimates the magnitude of the ozone mixing ratio, but matches the gradient with altitude well.

[42] Overall, the agreement between the 3-D CTM and the observation is quite good considering the CTM is constrained only by large-scale chemistry and meteorology. This demonstrates that a global model can reasonably represent such an event. It also demonstrates that the anomalous ozone profile is a product of the large-scale circulation. Further quantitative analysis, such as the frequency of these events, is beyond the scope of the present discussion but will be continued in a future study.

5. Conclusions

[43] Extensive simultaneous, in situ measurements in the UT/LS over a large region of the midlatitudes and subtropics has allowed for the characterization of the composition of the air mass in summer 2002. In July 2002, a large and persistent quasi-stationary anticyclone established a meridional flow pattern in the UT/LS. This was effective in transporting high-latitude air into the subtropics producing an altitude localized region of elevated ozone abundance between the tropopause and ~410 K. Both the tracer correlations and back trajectory calculations support this result. The extent of this layered structure covered over 12° latitude and was still observable near the tropics at 12°N.

[44] The general features of the ozone distribution are reproduced in a 3-D chemical transport model simulation. This indicates that these profiles are not a localized occurrence but a result of large-scale transport. Ultimately, improved model calculations constrained by extensive in situ observations will allow for a more quantitative evaluation of the larger-scale features and the implications for long-range transport of chemicals (and particulates) throughout the UT/LS. In particular, there exists a crucial need for a more complete understanding of the potential long-term global impacts involving the transport of midlatitude tropospheric pollutants into the tropical upwelling region.

[45] **Acknowledgments.** We wish to thank D. Kinnison and G. Brasseur for the MOZART version 3 CTM simulation results. We also acknowledge the dedicated efforts of all the NASA WB-57F pilots and crew during the CRYSTAL-FACE campaign. The authors also thank two anonymous reviewers for helpful comments on improving the manuscript.

References

Avallone, L. M., and M. J. Prather, Photochemical evolution of ozone in the lower stratosphere, *J. Geophys. Res.*, *101*, 1457–1461, 1996.

Brasseur, G. P., D. A. Hauglustaine, S. Walters, P. J. Rasch, J.-F. Muller, C. Granier, and X. X. Tie, MOZART, a global chemical transport model for ozone and related chemical tracers: 1. Model description, *J. Geophys. Res.*, *103*, 28,265–28,289, 1998.

Dunkerton, T. J., Evidence of meridional motion in the summer lower stratosphere adjacent to monsoon regions, *J. Geophys. Res.*, *100*, 16,675–16,678, 1995.

Fahey, D. W., et al., In situ observations of NO_y, O_3 and the NO_y/O_3 ratio in the lower stratosphere, *Geophys. Res. Lett.*, *23*, 1653–1656, 1996.

Flocke, F., et al., An examination of chemistry and transport processes in the tropical lower stratosphere using observations of long-lived and short-lived compounds obtained during STRAT and POLARIS, *J. Geophys. Res.*, *104*, 26,625–26,642, 1999.

Forster, P. M. de F., and K. P. Shine, Radiative forcing and temperature trends from stratospheric ozone changes, *J. Geophys. Res.*, *102*, 10,841–10,855, 1997.

Herman, R. L., et al., Tropical entrainment time scales inferred from stratospheric N_2O and CH_4 observations, *Geophys. Res. Lett.*, *25*, 2781–2784, 1998.

Houghton, J. T., et al. (Eds.), *Climate Change 2001: The Scientific Basis, Contribution of Working Group I to the Third Assessment Report of the International Panel on Climate Change (IPCC)*, pp. 349–416, Cambridge Univ. Press, New York, 2001.

Hudson, P. K., et al., Comparison of biomass burning plumes observed during the ITCT-2K2 and CRYSTAL-FACE missions, paper presented at Spring 2003 European Geophysical Union Meeting, Eur. Geophys. Soc., Nice, France, 2003.

Kelly, K. K., et al., Dehydration in the lower Antarctic stratosphere during late winter and early spring 1987, *J. Geophys. Res.*, *94*, 11,317–11,358, 1989.

Kley, D., et al., Transport of water through the tropical tropopause, *Geophys. Res. Lett.*, *9*, 617–620, 1982.

Lacis, A. A., D. J. Wuebbles, and J. A. Logan, Radiative forcing of climate by changes in the vertical distribution of ozone, *J. Geophys. Res.*, *95*, 9971–9981, 1990.

Logan, J. A., Trends in the vertical distribution of ozone: An analysis of ozonesonde data, *J. Geophys. Res.*, *99*, 25,553–25,585, 1994.

Logan, J. A., An analysis of ozonesonde data for the lower stratosphere: Recommendations for testing models, *J. Geophys. Res.*, *104*, 16,151–16,170, 1999.

Murphy, D. M., et al., Reactive nitrogen and its correlation with ozone in the lower stratosphere and upper troposphere, *J. Geophys. Res.*, *98*, 8751–8773, 1993.

Newman, P. A., D. W. Fahey, W. H. Brune, and M. J. Kurylo, Photchemistry of ozone loss in the Arctic region in summer (POLARIS), *J. Geophys. Res.*, *104*, 26,481–26,495, 1999.

Oltmans, S. J., H. Vömel, D. J. Hofmann, K. H. Rosenlof, and D. Kley, The increase in stratospheric water vapor from balloonborne frostpoint hygrometer measurements at Washington, D.C. and Boulder, Colorado, *Geophys. Res. Lett.*, *27*, 3453–3457, 2000.

Plumb, R. A., and M. K. W. Ko, Interrelationships between mixing ratios of long-lived stratospheric constituents, *J. Geophys. Res.*, *97*, 10,145–10,156, 1992.

Postel, G. A., and M. H. Hitchman, Climatology of Rossby wave breaking along the subtropical tropopause, *J. Atmos. Sci.*, *55*, 359–373, 1999.

Proffitt, M. H., and R. L. McLaughlin, Fast-response dual-beam UV-absorption ozone photometer suitable for use in stratospheric balloons, *Rev. Sci. Instrum.*, *54*, 1719–1728, 1983.

Proffitt, M. H., J. J. Margitan, K. K. Kelly, M. Loewenstein, J. R. Podolske, and K. R. Chan, Ozone loss in the Arctic polar vortex inferred from high altitude aircraft measurements, *Nature*, *347*, 31–36, 1990.

Proffitt, M. H., K. Aikin, A. F. Tuck, J. J. Margitan, C. R. Webster, G. C. Toon, and J. W. Elkins, Seasonally averaged ozone and nitrous oxide in the Northern Hemisphere lower stratosphere, *J. Geophys. Res.*, *108*(D3), 4110, doi:10.1029/2002JD002657, 2003.

Ramaswamy, V., M. D. Schwarzkopf, and K. P. Shine, Radiative forcing of climate from halocarbon-induced stratospheric ozone loss, *Nature*, *355*, 810–812, 1992.

Reid, S. J., and G. Vaughan, Lamination in ozone profiles in the lower stratosphere, *Q. J. R. Meteorol. Soc.*, *117*, 825–844, 1991.

Richard, E. C., K. K. Kelly, R. H. Winkler, R. Wilson, T. L. Thompson, R. J. McLaughlin, A. L. Schmeltekopf, and A. F. Tuck, A fast-response near-infrared tunable diode laser absorption spectrometer for in situ measurements of CH_4 in the upper troposphere and lower stratosphere, *Appl. Phys. B*, *75*, 183–194, 2002.

Ridley, B. A., J. G. Walega, J. E. Dye, and F. E. Grahek, Distributions of NO, NO_x, NO_y, and O_3 to 12 km altitude during the summer monsoon season over New Mexico, *J. Geophys. Res.*, *99*, 25,519–25,534, 1994.

Rood, R. B., A. R. Douglass, M. C. Cerniglia, L. C. Sparling, and J. E. Nielsen, Seasonal variability of middle-latitude ozone in the lowermost stratosphere derived from probability distribution functions, *J. Geophys. Res.*, *105*, 17,793–17,805, 2000.

Rosenlof, K. H., et al., Stratospheric water vapor increases over the past half century, *Geophys. Res. Lett.*, *28*, 1195–1199, 2001.

Russell, J. M., L. L. Gordley, J. H. Park, S. R. Drayson, D. H. Hesketh, R. J. Cicerone, A. F. Tuck, J. E. Frederick, J. E. Harries, and P. J. Crutzen, The Halogen Occulation Experiment, *J. Geophys. Res.*, *98*, 10,777–10,797, 1993.

Solomon, S., R. R. Garcia, and F. Stordal, Transport processes and ozone perturbations, *J. Geophys. Res.*, *90*, 12,981–12,989, 1985.

Stolarski, R. S., et al., 1995 scientific assessment of the atmospheric effects of stratospheric aircraft, *NSA Ref. Publ.*, *1381*, Nov. 1995.

Strahan, S. E., M. Lowenstein, J. R. Podolske, W. L. Starr, K. R. Chan, M. H. Proffitt, and K. K. Kelly, Correlation of N_2O and ozone in the southern polar vortex during the Airborne Antarctic Ozone Experiment, *J. Geophys. Res.*, *94*, 16,749–16,756, 1989.

Thompson, T. L., and K. H. Rosenlof, Accuracy and precision of the NOAA Aeronomy Laboratory pressure temperature instrument on the NASA WB-57F, paper presented at CRYSTAL-FACE Science Team Meeting, NASA, Salt Lake City, Utah, 2003. (Available at http://cloud1.arc.nasa.gov/crystalface/presentations_files/1-34_Rosenlof&Thompson.pdf)

Trepte, C. R., R. E. Veiga, and M. P. McCormick, The poleward dispersal of Mount Pinatubo volcanic Aerosol, *J. Geophys. Res.*, *98*, 18,563–18,573, 1993.

Tuck, A. F., et al., The Brewer-Dobson circulation in the light of high altitude in situ aircraft observations, *Q. J. R. Meteorol. Soc.*, *123*, 1–69, 1997.

Vaughan, G., and C. Timmis, Transport of near-tropopause air into the lower midlatitude stratosphere, *Q. J. R. Meteorol. Soc.*, *124*, 1559–1578, 1998.

Volk, C. M., et al., Quantifying transport between the tropical and mid-latitude lower stratosphere, *Science*, *272*, 1763–1768, 1996.

Weinheimer, A. J., J. G. Walega, B. A. Ridley, G. W. Sachse, B. E. Anderson, and J. E. Collins, Stratospheric NO_y measurements on the NASA DC-8 during AASE-II, *Geophys. Res. Lett.*, *20*, 2563–2566, 1993.

Weinheimer, A. J., J. G. Walega, B. A. Ridley, B. L. Gary, D. R. Blake, N. J. Blake, F. S. Rowland, G. W. Sachse, B. E. Anderson, and J. E. Collins, Meridional distributions of NO_x, NO_y, and other species in the lower stratopshere and upper troposphere during AASEII, *Geophys. Res. Lett.*, *21*, 2583–2586, 1994.

World Meteorological Organization, Scientific assessment of ozone depletion: 2002, *Global Ozone Res. and Monit. Proj. Rep. 47*, Geneva, Switzerland, 2003.

K. C. Aikin, E. A. Ray, and E. C. Richard, Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO 80309, USA. (richard@al.noaa.gov)

A. Gettelman, National Center for Atmospheric Research, Boulder, CO 80303, USA.

D. Knapp, D. Montzka, B. Ridley, and A. Weinheimer, Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, CO 80303, USA.

K. H. Rosenlof and T. L. Thompson, Aeronomy Laboratory, National Ocean and Atmospheric Administration, Boulder, CO 80305, USA.